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N. E. R. Zimmermann,* S. Jakobtorweihen, E. Beerdsen, B. Smit, and F. J. Keil: In-Depth Study of the Influence of Host−Framework Flexibility on the Diffusion of Small Gas Molecules in One-Dimensional Zeolitic Pore Systems

Page 17370. Figure 7: Because of an error in the analysis, the transition state theory diffusion coefficients, D_S^{TST}, of methane in AFI are too large by a factor of 3. As $\kappa = D_S^{MSD}/D_S^{TST}$, the dynamical correction factors are too small by a factor of 3. However, these factors cancel out exactly for the corresponding enhancement values (Figure 7d), which are the main focus of the paper.

Table 5: D_S^{TST} and thus κ are not correct because the histogram sampling assumed a symmetry that is not applicable to the LTL and MTW pores. Therefore, new simulations had to be conducted. The results are detailed in Table 1 (see below).

The new simulation results led to a +5.8% and −0.3% reduction of the diffusion barrier for LTL using the modified Demontis and Nicholas force field, respectively.

Page 17380, second column, 18th last line: "larger diffusion coefficient" should read "larger TST diffusion coefficient".

Page 17380, second column, 14th last line: "decrease" should read "increase".

Since the changes in the LTL and MTW enhancement values are small, *all* general conclusions of the paper remain the same. The absolute values of the dynamical correction factors at zero loading (almost all are larger than unity) deserve some comments.

In the framework of dynamically corrected transition state theory, the dynamical correction factor is, by definition, $0 \leq \kappa \leq 1$ because it is identified as the plateau of the reactive flux correlation function (RFCF), $\kappa(t)$:[1]

TABLE 1: TST Diffusivities and Dynamical Correction Factors; Compare with Table 5 in the Original Article

		rigid		mod. Demontis		Nicholas	
	θ [1/UC]	D_S^{TST} [10^{-8} m^2/s]	κ	D_S^{TST} [10^{-8} m^2/s]	κ	D_S^{TST} [10^{-8} m^2/s]	κ
LTL	0	0.460	1.007	0.518	0.946	0.462	1.004
	2	0.667	0.610	0.722	0.650	0.662	0.656
	7	1.336	0.139	1.449	0.154	1.290	0.177
MTW	0	1.913	1.402	1.871	1.657	2.053	1.228
	2	1.757	0.449	1.697	0.497	1.957	0.381
	7	1.221	0.039	1.423	0.059	1.302	0.052

Figure 9: The enhancement values of D_S^{TST} and κ for LTL and MTW changed only slightly due to the new simulation results; see Figure 1 below.

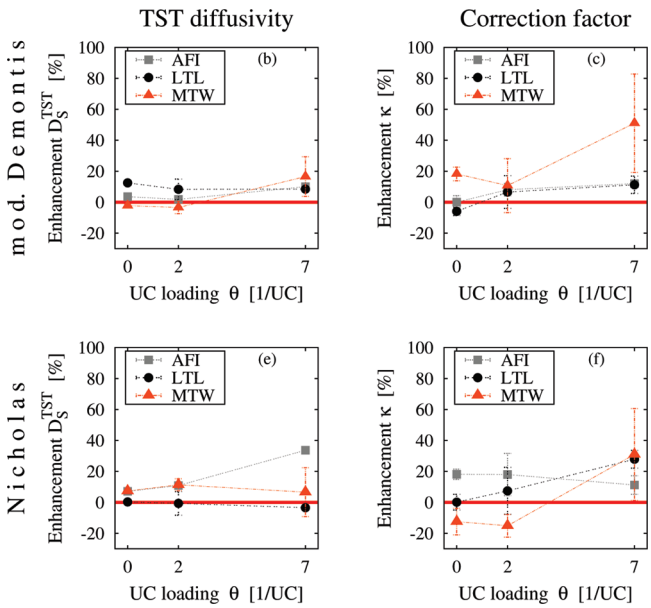

Figure 1. Enhancement factors for TST diffusivity and dynamical correction factor; compare with Figure 9 in the original article.

Table 6: The relative reduction of the free-energy barriers was computed as $(\Delta F^R - \Delta F^F)/\Delta F^R$; not as $(\Delta F^F - \Delta F^R)/\Delta F^R$.

$$\kappa(t) = \frac{\langle \dot{q}(0) \cdot H[q(t) - q^{\ddagger}] \cdot \delta[q(0) - q^{\ddagger}] \rangle}{\langle 0.5 \cdot |\dot{q}(0)| \rangle} \quad (1)$$

The RFCF is usually obtained from a different simulation than the free-energy profile and we have recently shown that *this* κ strictly equals unity for methane in rigid AFI at zero loading.[2] In contrast, we have, in the flexibility study, used the same simulation to obtain the mean-squared displacement and the free-energy profile and the dynamical correction factor was then indirectly determined by the ratio of the MSD-to-TST diffusivity. In doing so, the correction factor probes not only short-term correlations, as the reactive flux function does, but also long-term (kinetic) correlations. The latter type occurs when the free-energy barrier is relatively small (on the order of 1 k_BT), as is the case for methane in the channel-type zeolites studied here. Because of the small barrier, a particle with sufficiently large momentum that has just jumped into a neighboring cage will tend to immediately jump into even the next cage to produce a cascade of correleated jumps in the same direction. Note that this particle memory was, for example, systematically investigated by Kärger et al. for ethane molecules diffusing in silicalite.[3] The dynamical correction factor, as calculated in this work, should therefore rather be labeled "correlation factor" to underline that it accounts for a variety of correlations.

References and Notes

(1) Chandler, D. *J. Chem. Phys.* **1978**, *68*, 2959–2970.
(2) Zimmermann, N. E. R.; Smit, B.; Keil, F. J. *J. Phys. Chem. C* **2010**, *114*, 300–310.
(3) Kärger, J.; Demontis, P.; Suffritti, G. B.; Tilocca, A. *J. Chem. Phys.* **1999**, *110*, 1163–1172.

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